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#### DETAILED DESCRIPTION

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[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to all solid-state lithium ion batteries and the manufacture approach of those, and relates to all the solid-state lithium ion batteries that improved the solid electrolyte layer which covers the periphery of an anode plate especially, and the manufacture approach of those.

[0002]

[Description of the Prior Art] Memory and a processor are carried and the IC card which has informational storage and an informational processing facility is developed as a money card, a driver's license, etc. It is the thin thing which may be carried in a thin IC card as a cell which there are a thing of a cell loading mold and a thing of a cell a non-carried mold as this kind of an IC card, among these is used for the IC card of a cell loading mold, and since it is what people moreover carry, safety needs to be high.

[0003] As a cell with high safety, all solid-state lithium ion batteries with the easy closure are known for such a thin shape without using liquid.

[0004] There are some which were considered as the configuration as shown in drawing 4 as an example in all these solid-state lithium ion batteries. That is, invagination of the anode plate active material layer 2 is carried out on the substrate 1 which consists of stainless steel of all these solid-state lithium ion batteries etc., and serves as an anode plate, and the periphery of this anode plate active material layer 2 is covered with the solid electrolyte layer 3. Moreover, on this solid electrolyte layer 3, the laminating of the negative-electrode active material layer 4 is carried out, and the periphery of this negative-electrode active material layer 4 is covered with the negative electrode 5. furthermore, the periphery of said solid electrolyte layer 3 and said negative electrode 5 -- moisture-proof -- business -- it is covered with the enveloping layer 6.

[0005]

[Problem(s) to be Solved by the Invention] By the way, as a solid electrolyte layer 3 of all the account solid-state lithium ion batteries mentioned above, it is  $\text{Li}_{x}\text{Al}_{y}\text{Ti}_{z}(\text{PO}_4)_3$ . This  $\text{Li}_{x}\text{Al}_{y}\text{Ti}_{z}(\text{PO}_4)_3$   $\text{TiO}_2$   $\text{ZrO}_2$  Although the ceramic material which solidified the added melting mixture is known Since this ceramic material is weak, if it is difficult to form sheet metal and the thin film 100 micrometers or less of thickness and it forms this ceramic material in the plate with comparatively thick thickness suitable for the sources for vacuum evaporation, such as a target, conversely There was a possibility that internal strain might arise and it might be generated [ crack ].

[0006] Therefore, thickness was not able to form the solid electrolyte layer which has high ion conductivity very thin moreover.

[0007] This invention aims at offering all the solid-state lithium ion batteries that have the solid electrolyte layer in which thickness has high ion conductivity very thin moreover, and the manufacture approach of those in view of such a point.

[0008]

[Means for Solving the Problem] A solid electrolyte layer has the description of all the solid-state lithium ion batteries of this invention which relates to claim 1 in order to attain the purpose mentioned above in the point of making the molding which consists of at least two kinds of fine particles of a lithium compound, a metallic oxide, and the sulfuration compounds coming to disperse

as the source for vacuum evaporationo. And since a solid electrolyte layer is formed as two or more sorts of fine particles mixed by homogeneity by having adopted such a configuration fix mutually, thickness can consider as the solid electrolyte layer which has high ion conductivity very thin moreover.

[0009] The description of all the solid-state lithium ion batteries of this invention concerning claim 2 is that it set particle size of the fine particles of a lithium compound, a metallic oxide, or a sulfuration compound to 0.5-300 micrometers. And by having adopted such a configuration, two or more sorts of fine particles can be mixed to homogeneity, and it can fix good mutually.

[0010] The description of all the solid-state lithium ion batteries of this invention concerning claim 3 is that it should grind the ceramic about the fine particles for forming a solid electrolyte layer. And since what was crystallized by having adopted such a configuration is ground and used, two or more sorts of fine particles can be further mixed to homogeneity, and it can fix good mutually.

[0011] The description of the manufacture approach of all the solid-state lithium ion batteries of this invention concerning claim 4 mixes at least two kinds of fine particles of a lithium compound , a metallic oxide , and the sulfuration compounds , makes heat and a pressure act on this mixture , casts them , and is that this molding was dispersed as the source for vacuum evaporationo , and it formed the solid electrolyte layer . And thickness can form the solid electrolyte layer which has high ion conductivity very thin moreover by having adopted such a configuration.

[0012] The description of the manufacture approach of all the solid-state lithium ion batteries of this invention concerning claim 5 is that formed the ceramic including at least two kinds in a lithium compound, a metallic oxide, and a sulfuration compound, ground this ceramic and formed fine particles, and made heat and a pressure act on these fine particles, it cast, this molding was dispersed as the source for vacuum evaporationo, and it formed the solid electrolyte layer. And by having adopted such a configuration, two or more sorts of fine particles can be further mixed to homogeneity, it can be made to be able to fix mutually good, and a solid electrolyte layer can be formed.

[0013]

[Embodiment of the Invention] Although this invention is applicable to the configuration of all the solid-state lithium ion batteries shown in drawing 4 mentioned above, it is applicable also to the configuration shown in drawing 3 .

[0014] Then, the configuration of all the solid-state lithium ion batteries shown in this drawing 3 is explained first.

[0015] That is, on the substrate 1 of all these solid-state lithium ion batteries, an anode plate 7 and a negative electrode 5 separate spacing, the laminating is mutually carried out, and the laminating of the anode plate active material layer 2 with a flat-surface dimension smaller than an anode plate 7 is carried out on said anode plate 7. Moreover, the periphery of said anode plate 7 and the anode plate active material layer 2 is covered with the solid electrolyte layer 3 except for said a part of anode plate 7. Furthermore, the one section each of said negative electrode 5 and said solid electrolyte layer 3 is covered with the negative-electrode active material 4. further -- again -- the one section each of said anode plate 7 and a negative electrode 5, and the periphery of said negative-electrode active material 4 -- moisture-proof -- business -- it is covered with the enveloping layer 6.

[0016] as said anode plate active material layer 2 -- V2O<sub>x</sub> (x=1-5), Li<sub>x</sub>MnO<sub>y</sub>; Li<sub>x</sub>CoO<sub>y</sub>, LiNiO<sub>y</sub>, Li<sub>x</sub>FeO<sub>2</sub>, Li<sub>x</sub>TiO<sub>y</sub>, Li<sub>x</sub>ScO<sub>y</sub>, and Li<sub>x</sub>YO<sub>y</sub> etc. -- it is used.

[0017] moreover, Li<sub>x</sub>Si<sub>y</sub>O<sub>z</sub> which are the ingredient and the various graphite which made Li, Li-aluminum, and aluminum diffuse Li, carbon and these analogs, or a stannic-acid ghost compound as said negative-electrode active material layer 4 etc. -- it is used.

[0018] Furthermore, as said substrate 1, what vapor-deposited metals, such as Cu, nickel, V, aluminum, Pt, and Au, is used on films, such as a polyamide or polyimide, PET and PPS, and polypropylene, or glass and silicon plates, such as stainless steel with a thickness of 10-500 micrometers, and Cu, nickel, V, Au, Pt, aluminum.

[0019] In all the solid-state lithium ion batteries mentioned above, said solid electrolyte layer 3 is the following, and is made and formed.

[0020] namely, -- as the solid electrolyte layer 3 -- first -- as a target ingredient -- Li<sub>x</sub> M (M=PO<sub>4</sub> --) S and SiO<sub>4</sub> etc. -- etc. -- a lithium compound and aluminum2 -- O<sub>3</sub> and TiO<sub>2</sub> -- The molding which

consists of at least two kinds of fine particles of the sulfuration (M=aluminum, Ti, Zr, Fe and Ga, In X=1-5) compounds of metallic oxides, such as SiO<sub>2</sub>, ZrO<sub>2</sub>, Fe<sub>2</sub>O<sub>3</sub>, Ga<sub>2</sub>O<sub>3</sub>, and B-2s In [ O<sub>3</sub> and ] 2O<sub>3</sub>, M<sub>x</sub>S, etc. is formed.

[0021] When an example of an ingredient for this is given, it is aluminum 2O<sub>3</sub> and TiO<sub>2</sub> considering Li<sub>3</sub>PO<sub>4</sub> as 60-mol % and a metallic oxide as a lithium compound. About aluminum 2O<sub>3</sub>, it is six-mol% and TiO<sub>2</sub>. If it attaches, 34-mol % of fine particles are mixed mutually. As for the powder diameter of the fine particles of each ingredient, at this time, it is desirable that it is for 0.5-300 micrometers. Although a powder diameter is so good that it is small in order for this to make homogeneity distribute each ingredient, it is because a to some extent big particle size is needed in order to form molding.

[0022] In addition, before forming the fine particles mentioned above, a ceramic may be formed with the same ingredient at the pressure of 10kg/cm<sup>2</sup> - 20 t/cm<sup>2</sup>, and the temperature of 0 degree C - 1000 degrees C, and this ceramic may be ground so that it may become fine particles with a particle size of 0.5-300 micrometers. Thus, further homogenization can be enabled by forming a ceramic beforehand.

[0023] And after mixing these ingredients to homogeneity, as it is shown in this ingredient at drawing 1, they are 10kg/cm<sup>2</sup> - 20 t/cm<sup>2</sup>. The molding 10 as a solid target can be formed as the whole with a thickness of 5mm for the diameter of 3 inches by making a pressure act in a 0 degree C - about 1000 degrees C temperature requirement, and making each fine particles A, B, and C fix mutually.

[0024] Below, drawing 2 explains the vacuum evaporationo equipment for carrying out the laminating of the solid electrolyte layer 3 on the anode plate active material layer 2.

[0025] Vacuum evaporationo equipment 11 has the sealing casing 12, the substrate 1 with which the laminating of the anode plate which omitted illustration, cathode, the anode plate active material layer, etc. was carried out is supported by the upper part in this sealing casing 12, and the part of vacuum evaporationo needlessness is covered with the mask which is not illustrated so that a solid electrolyte layer can be vapor-deposited only on the anode plate active material layer of this substrate 1.

[0026] The boat 15 heated by the electron beam gun 14 excited by the external EB power source 13 is arranged by the lower part in said sealing casing 12, and said molding 10 as the source for vacuum evaporationo, i.e., a target, is arranged by this boat 15.

[0027] Moreover, while the oxygen radical generator 16 which supplies an oxygen radical towards said substrate 1 is arranged towards the interior from the exterior of said sealing casing 12 and an oxygen supply 17 is connected to this oxygen radical generator 16, RF generator 18 is arranged. Furthermore, in said sealing casing 12, the shutter 19 with which the upper and lower sides are divided partially is arranged.

[0028] And by supplying the oxygen radical as assistance towards a substrate 1 with the oxygen radical generator 16, while supplying the electron beam generated from the electron beam gun 14 excited by the power supplied from the EB power source 13 to a boat 15, evaporating said molding 10 as a target and carrying out EB vacuum evaporationo, a solid electrolyte layer is formed so that the anode plate active material layer on a substrate 1 may be covered.

[0029] Thus, since each component is homogenized, the formed anode plate active material layer can make thickness thin with 0.2-10 micrometers, and, moreover, can form the good solid electrolyte layer of ion conductivity.

[0030] Therefore, all the solid-state lithium ion batteries of this operation gestalt which has such a solid electrolyte layer have a long charge-and-discharge cycle life, and current density can be made high.

[0031] As a result of performing the charge and discharge test of all the solid-state lithium ion batteries of this operation gestalt and measuring a cell property, as shown in the following table, it is 2 1cm. Even if it passed the current of 200micro [ of hits ] A, the big change in cell capacity was not seen.

[0032]

固定電流 ( $\mu\text{A}/\text{cm}^2$ )	電池容量 ( $\mu\text{A h r}/\text{cm}^2$ )	$I = 10 \mu\text{A}/\text{cm}^2$ 時を 100%とした場合
10	44.0	100%
50	40.1	91%
100	38.5	88%
200	32.2	79%

In addition, this invention is not limited to the gestalt of operation mentioned above, and various modification is possible for it if needed. For example, this invention is not limited to EB vacuum deposition mentioned above, and can apply other membrane formation approaches, such as a spatter and the laser ablation method.

[0033]

[Effect of the Invention] As explained above, according to this invention, thickness can offer all the solid-state lithium ion batteries that have the solid electrolyte layer which has high ion conductivity very thin moreover, and the manufacture approach of those.

[0034] That is, since the molding which a solid electrolyte layer becomes from at least two kinds of fine particles of a lithium compound, a metallic oxide, and the sulfuration compounds is dispersed as the source for vacuum evaporationo and a solid electrolyte layer is formed as two or more sorts of fine particles mixed by homogeneity fix mutually, thickness can use all the solid-state lithium ion batteries of this invention as the solid electrolyte layer which has high ion conductivity very thin moreover.

[0035] Moreover, 0.5-300 micrometers, then two or more sorts of fine particles can be mixed to homogeneity, and the particle size of the fine particles of a lithium compound, a metallic oxide, or a sulfuration compound can be fixed good mutually.

[0036] Furthermore, since the thing which ground the ceramic for the fine particles for forming a solid electrolyte layer, then the thing crystallized once are ground and used, two or more sorts of fine particles can be further mixed to homogeneity, and it can fix good mutually.

[0037] On the other hand, since the manufacture approach of all the solid-state lithium ion batteries of this invention mixed at least two kinds of fine particles of a lithium compound, a metallic oxide, and the sulfuration compounds, made heat and a pressure act on this mixture, cast them, dispersed this molding as the source for vacuum evaporationo and formed the solid electrolyte layer, it can form the solid electrolyte layer in which thickness has high ion conductivity very thin moreover.

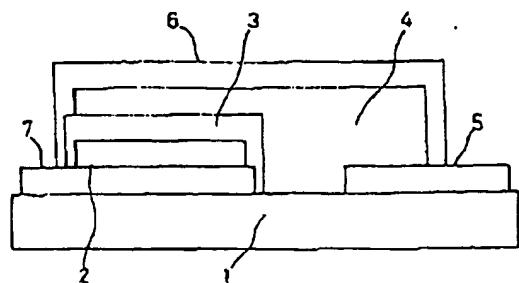
[0038] Moreover, if a ceramic including at least two kinds in a lithium compound, a metallic oxide, and a sulfuration compound is formed, this ceramic is ground, fine particles are formed, make heat and a pressure act on these fine particles, it casts, this molding is dispersed as the source for vacuum evaporationo and a solid electrolyte layer is formed, two or more sorts of fine particles can be further mixed to homogeneity, it can be made to be able to fix mutually good, and a solid electrolyte layer can be formed.

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[Translation done.]

Drawing selection **drawing 3**

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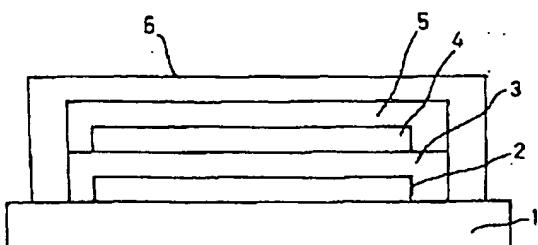


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Drawing selection drawing 4



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